Giant Ambipolar Diffusion Constant of *n-i-p-i* Doping Superlattices

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(Received 5 November 1990)

The analytical expression for the ambipolar in-plane diffusion constant of excess carriers in *n-i-p-i* doping superlattices is derived. Shockley-Haynes–type experiments quantitatively confirm the predicted huge enhancement of the diffusion constant (in the tested case by factors of about 300) and the long diffusion lengths.

PACS numbers: 73.50.Pz, 73.20.Dx, 73.50.Gr

n-i-p-i crystals[1,2] represent a unique system to study the dynamic behavior of strongly correlated electron and hole plasmas. Contrary to the bulk,[3] where Coulomb interaction is highly efficient in keeping the electron-hole plasma neutral and where short recombination[4] lifetimes also prohibit diffusion over large distances, the strong built-in space-charge fields in *n-i-p-i* systems fully separate electrons and holes from each other on the scale of the superlattice period.[1,2] Therefore, the electrostatic repulsion between carriers of the same type is no longer compensated by the attractive interaction between carriers of opposite sign. This results in strong space-charge–induced fields of opposite sign for both types of carriers, which enormously enhance their ambipolar diffusion. Moreover, the spatial separation suppresses electron-hole recombination to a large extent. Recent theoretical and experimental studies[5-7] have already indicated the existence of a new, extremely fast diffusive relaxation mechanism for locally generated charge carriers in such systems.

In this paper we first present a general derivation of the analytical expression for the diffusion constant in *n-i-p-i* systems.[5] The predicted huge value of the diffusion constant is then verified by a Shockley-Haynes–type experiment.[3] In addition, we provide the experimental proof for the dramatically increased value of the diffusion length in GaAs *n-i-p-i* systems. It is shown that even for diffusion over distances in the cm range recombination can be negligible.

For the derivation of the diffusion constant, consider the steady-state situation of a large *n-i-p-i* sample which is continuously illuminated at a small spot. Recombination shall take place only via externally connected selective *n*- and *p*-type contacts far away from the excitation point. The continuum equations for the electron and hole currents $\mathbf{j}_n$ and $\mathbf{j}_p$ are

$$\mathbf{j}_n(\mathbf{r}) = \mu_n n \nabla\Phi_n(\mathbf{r}) , \tag{1}$$

$$\mathbf{j}_p(\mathbf{r}) = \mu_p p \nabla\Phi_p(\mathbf{r}) , \tag{2}$$

where μ_n, μ_p, n, p, Φ_n, and Φ_p are the mobilities, densities, and the respective quasi-Fermi-levels, and $\mathbf{r}=(x,y,z)$. Note that by using the gradient of the quasi-Fermi-level as the "driving force" for the current, both diffusion and drift currents are automatically included.

Since there is no external field in our case we have only a diffusion current. Its two components compensate each other such that the total current vanishes:

$$\mathbf{j}(\mathbf{r}) = \mathbf{j}_n(\mathbf{r}) + \mathbf{j}_p(\mathbf{r}) \equiv 0 . \tag{3}$$

This implies the following relation between the gradients of the electron and hole quasi-Fermi-levels:

$$\nabla\Phi_p(\mathbf{r}) = -[\sigma_n(\mathbf{r})/\sigma_p(\mathbf{r})]\nabla\Phi_n(\mathbf{r}) . \tag{4}$$

This relation is also valid in bulk material and reflects the fact that, in the ambipolar case, the diffusion of carriers with different mobility results in the buildup of space-charge fields.

The ambipolar diffusion constant D_a is defined by

$$\mathbf{j}_n(\mathbf{r}) = eD_a \nabla n(\mathbf{r}) = -\mathbf{j}_p(\mathbf{r}) = eD_a \nabla p(\mathbf{r}) . \tag{5}$$

For nondegenerate semiconductors the local electron and hole densities and their gradients can be expressed by the effective electron and hole densities of states, N_c and N_v, and the difference between the respective quasi-Fermi-levels and the band-edge energies $E_c(\mathbf{r})$ and $E_v(\mathbf{r})$:

$$n(\mathbf{r}) = N_c e^{-[E_c(\mathbf{r})-\Phi_n(\mathbf{r})]/kT} , \tag{6}$$

$$p(\mathbf{r}) = N_v e^{-[\Phi_p(\mathbf{r})-E_v(\mathbf{r})]/kT} , \tag{7}$$

$$\nabla n(\mathbf{r}) = -n(\mathbf{r})(\nabla E_c - \nabla\Phi_n)\frac{1}{kT} , \tag{8}$$

$$\nabla p(\mathbf{r}) = -p(\mathbf{r})(\nabla\Phi_p - \nabla E_v)\frac{1}{kT} . \tag{9}$$

In *n-i-p-i* crystals, however, the local carrier densities are determined by the local quasi-Fermi-level differences $\Phi_{np} = \Phi_n - \Phi_p$.[2] Thus, the gradients of the electron and hole densities are now determined by the relation

$$\nabla n(\mathbf{r}) = \nabla p(\mathbf{r}) = \frac{\partial n(\rho)}{\partial\Phi_{np}}(\nabla_\rho\Phi_n - \nabla_\rho\Phi_p) \tag{10}$$

which differs completely from Eqs. (8) and (9). In Eq. (10) we have replaced the two-dimensional sheet carrier densities by averaged bulk values, obtained by division of $n^{(2)}(\mathbf{r})$ and $p^{(2)}(\mathbf{r})$ by the superlattice period d. As diffusion in the superlattice direction z is negligible, we have also introduced the two-dimensional in-plane vector $\boldsymbol{\rho}=(x,y)$ and the two-dimensional in-plane gradient $\nabla_\rho=(\partial/\partial x,\partial/\partial y)$. We note that the expression $d^{-1}\partial n^{(2)}/\partial\Phi_{np}$ multiplied by e^2 can be interpreted as the (three-dimensional) capacitance density $C_{np}(\Phi_{np})$ of the n-i-p-i system.[8]

The relation $n(\Phi_{np})$ is closely linked to the tunable effective band gap $E_g^{\text{eff}}(n)$ in n-i-p-i crystals. In the extreme cases of uniformly[2] or δ-doped[1,9] n-i-p-i's one has a parabolic or linear dependence, respectively. In Fig. 1 these dependences are shown for the specific case of p-type n-i-p-i semimetals (defined as n-i-p-i structures with finite electron and hole concentrations $p>n$ in the ground state $\Phi_{np}=0$). Note that $p(\Phi_{np})-n(\Phi_{np})=\text{const}$, because of the requirement of (macroscopic) neutrality of the n-i-p-i structure.[1,2] For $\Phi_{np}\le\Phi_{np}^{\text{th}}$, i.e., below the "depletion threshold" Φ_{np}^{th}, the two-dimensional carrier density $n^{(2)}$ drops rapidly to zero and our simplified considerations are no longer applicable.

If we use Eq. (4) to eliminate $\nabla_\rho\Phi_p$ in Eq. (10) we obtain

$$\nabla_\rho\Phi_n=\frac{1}{(1+\sigma_n/\sigma_p)}\frac{\partial\Phi_{np}}{\partial n}\nabla_\rho n(\mathbf{r})\,. \qquad (11)$$

Inserting Eq. (11) in (1) and (2) and comparing with (5) we obtain the expression for the ambipolar diffusion constant of n-i-p-i systems,

$$D_a^{nipi}=\frac{1}{e^2}\frac{\sigma_n\sigma_p}{\sigma_n+\sigma_p}\frac{\partial\Phi_{np}}{\partial n}\,, \qquad (12)$$

whereas with Eqs. (8) and (9) instead of (10) we reproduce the well-known formula for the ambipolar diffusion constant in bulk material,[3,10]

$$D_a^{\text{bulk}}=\frac{1}{e^2}\frac{\sigma_n\sigma_p}{\sigma_n+\sigma_p}\left(\frac{1}{n}+\frac{1}{p}\right)kT\,. \qquad (13)$$

It is well known [and can be seen easily from Eq. (13)] that in bulk semiconductors the values of the ambipolar diffusion constant lie in between those for electrons or holes,

$$D_p\equiv\mu_p\frac{kT}{e}<D_a^{\text{bulk}}<\mu_n\frac{kT}{e}\equiv D_n\,. \qquad (14)$$

From Eq. (12) we deduce the estimates

$$\mu_n n\frac{\partial\Phi_{np}}{\partial n}\frac{1}{2e}<D_a^{nipi}<\mu_n n\frac{\partial\Phi_{np}}{\partial n}\frac{1}{e} \quad \text{if } \mu_n n<\mu_p p\,, \qquad (15)$$

$$\mu_p p\frac{\partial\Phi_{np}}{\partial n}\frac{1}{2e}<D_a^{nipi}<\mu_p p\frac{\partial\Phi_{np}}{\partial n}\frac{1}{e} \quad \text{if } \mu_n n>\mu_p p\,. \qquad (16)$$

Obviously, $n\partial\Phi_{np}/\partial n$ and $p\partial\Phi_{np}/\partial n$ are always much larger than kT in any reasonable n-i-p-i structure, unless n or p approach zero. Using our example from Fig. 1, or by inserting other typical values for n, p, and $\partial n/\partial\Phi_{np}$, it is easy to verify that the "enhancement factors" for ambipolar diffusion $(n\partial\Phi_{np}/\partial n)/kT$ or $(p\partial\Phi_{np}/\partial n)/kT$ are typically of the order of 100.

In order to test this prediction we performed time-resolved measurements of the diffusion current caused by a short light pulse at a distance x from the measuring contacts. The diffusion distance was varied from 0.7 to 8.6 mm. The "half-period n-i-p-i" samples were grown by molecular-beam epitaxy with the following design:[11] semi-insulating GaAs substrate, 1000-nm p-doped GaAs ($n_A=5.5\times10^{16}$ cm^{-3}), 800-nm n-GaAs ($n_D=2.8\times10^{16}$ cm^{-3}). Lithographic techniques were employed for the structuring. Selective n- and p-type contacts[2] were fabricated by evaporating Au/Zn onto the p, and Ni/Sn onto the n layer. The width of the structure is 40 μm and its length is 8.7 mm (see inset of Fig. 2).

The sample was locally excited by short (30-ns) laser pulses of wavelength 780 nm and intensity 148 μW. The diameter of the excitation spot was 20 μm. The generated carriers recombined externally via resistor R_{np}, connecting two closely spaced selective n- and p-type contacts. The resistor R_{np} was chosen such that the external lifetime of the carriers[2] was much shorter than the internal n-i-p-i lifetime but, at the same time, was much longer than the diffusion times. The applied reverse bias was -3 and -1 V, respectively. The time dependence of the photovoltage $\Delta U_{np}(t)$ between two closely spaced measuring contacts has been measured by monitoring the voltage drop along the external resistor with a digital oscilloscope. The resulting curves $\Delta I_{np}(t)$ which are pro-

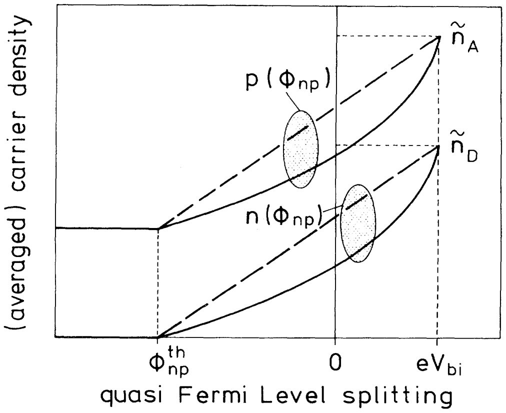

FIG. 1. Schematic diagram depicting the dependence of the (averaged) carrier concentrations n and p on the quasi-Fermi-level splitting $\Phi_{np}=eU_{np}$ for p-type n-i-p-i semimetals. Solid lines: n-i-p-i with $d_i=0$; dashed lines: δ-doped n-i-p-i.

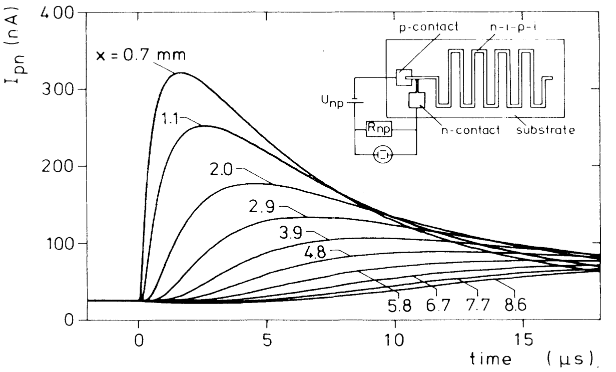

FIG. 2. Transient behavior of the recombination current between two closely spaced n- and p-type contacts. The diffusion distances were varied from 8.6 to 0.7 mm in steps of 0.9 mm. The inset indicates the sample geometry. The meander shape was used in order to enhance the diffusion length. The applied reverse bias U_{pn} was -3 V; the external resistor R_{np} was 1 MΩ.

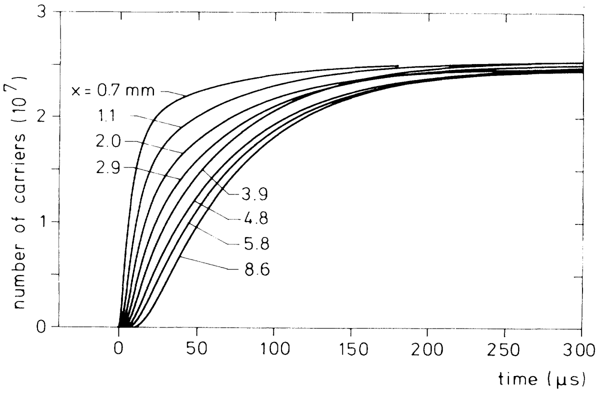

FIG. 3. Time integral $N(t) = (1/e)\int_0^t I_{np}(t')\,dt'$ of the recombination current for different diffusion distances. Note that even for a diffusion distance of 8.6 mm the total number of carriers which have arrived at the detection contacts has not significantly decreased.

portional to $\Delta n(t)$ are shown in Fig. 2 for different diffusion distances, 0.7 mm $\leq x \leq$ 8.6 mm; $R_{np} = 1$ MΩ and $U_{np} = -3$ V.

We note that diffusive transport over such large distances is impossible in bulk GaAs, since recombination limits the ambipolar diffusion length to values of $\Lambda_a^{\text{bulk}} < 100$ μm, even for very pure samples.[10] In n-i-p-i crystals, however, the long lifetimes result in negligible recombination even for distances which exceed the bulk diffusion lengths by several orders of magnitude. In fact, we can see from Fig. 3 that recombination is negligible even for our longest diffusion distances of 8.6 mm. This figure displays the time integrals of the I_{np} curves which are proportional to the number of carriers that actually have arrived at the time t at the contacts. One can easily see that this number is independent of the diffusion distance x even for $x = 8.6$ mm.

Because of the favorable quasi-one-dimensional geometry and the lack of recombination, the relaxation of the local excess charge density can be described by Ficks second law:

$$\frac{\partial \Delta n}{\partial t} = -D_a \frac{\partial^2 \Delta n}{\partial x^2} . \tag{17}$$

Its general solution for an infinite one-dimensional sample and a spatially and temporally δ-shaped excitation pulse is

$$\Delta n(x,t) = \frac{\Delta n_0}{(4\pi D_a^{nipi} t)^{1/2}} \exp\left[-\frac{x^2}{4 D_a^{nipi} t} \right] \tag{18}$$

(Δn_0 denotes the number of generated charge carriers per y-z cross section). Since $\Delta I_{np}(t) \propto \Delta n(t)$ a plot of $\ln(t^{1/2}\Delta I_{np})$ vs $x^2/4t$ should therefore reveal straight lines with the diffusion constant as the reciprocal slope. Figure 4 shows such a semilogarithmic plot of the data of Fig. 2. Independent of the diffusion distance, the curves, indeed, exhibit the same slope.

The slight deviation for distances ≤ 1 mm is due to the electrical setup of the experiment which restricts the minimum rise time to 600 ns. The reciprocal slopes of the curves of Fig. 4 yield an experimental value for the ambipolar diffusion constant of $(D_a^{nipi})^{\text{expt}} = 5000 \pm 300$ cm^2/s.

Since the electron and hole concentrations and mobilities of the sample are known,[11] the ambipolar diffusion constant can be calculated. Using the conventional bulk diffusion theory [Eq. (14)], a value of $D_a^{\text{bulk}} = 17$ cm^{2}s^{-1} is obtained for $U_{np} = -3$ V.

The giant value of the diffusion constant as deduced from experiment, which is more than 2 orders of magnitude above the corresponding bulk value, is in perfect

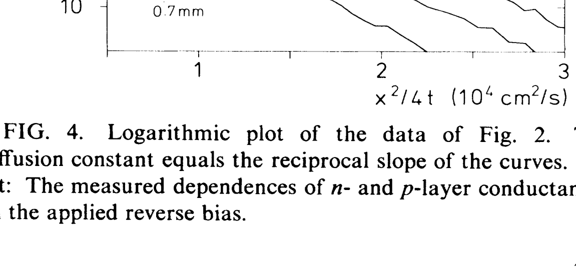

FIG. 4. Logarithmic plot of the data of Fig. 2. The diffusion constant equals the reciprocal slope of the curves. Inset: The measured dependences of n- and p-layer conductances on the applied reverse bias.

agreement with Eq. (12) for the ambipolar diffusion constant in n-i-p-i systems. Using this equation to calculate the diffusion constant from the measured dependences of n- and p-layer conductances on the applied reverse bias[11] a theoretical value of $(D_a^{nipi})^{\text{theor}} = 5500 \pm 1000$ cm^2/s is calculated, which is within the experimental uncertainty of the values for n, p, μ_n, and μ_p (see inset of Fig. 4).

As $\partial n/\partial \Phi_{np}$ and σ_n and σ_p are tunable quantities in a n-i-p-i crystal, the value of the diffusion constant can also be changed dynamically as a function of the applied reverse bias U_{pn}. For an applied reverse bias of -1 V instead of -3 V one calculates a reduced value of 4600 ± 800 cm^2/s. This reduced value of the diffusion constant has also been verified experimentally. Performing the same experiment and analysis as before we deduce $D_a^{nipi}(-1 \text{ V}) = 4400 \pm 400$ cm^{2}s^{-1}.

In summary, we have derived an analytical expression for the ambipolar diffusion constant in n-i-p-i systems. We have quantitatively verified our prediction of dramatically increased values of D_a^{nipi} as compared to the corresponding bulk values D_a^{bulk} as well as the dependence on the applied reverse bias, by Shockley-Haynes-type experiments. We have also demonstrated that diffusion over macroscopic distances in the cm range is possible in n-i-p-i systems without the occurrence of any significant recombination. Even at our maximum diffusion distance of nearly 1 cm recombination is negligible.

The fast diffusion has important implications for the application of n-i-p-i structures with selective n- and p-type contacts[2,5] in fast optoelectric devices. Even with lateral dimensions of 10 μm, a 3-dB frequency of 1 GHz becomes possible in GaAs n-i-p-i systems. Investigations on other related peculiarities of the in-plane transport, such as carrier propagation in the case of injection by spatially separated selective n- and p-type contacts, are in progress.

We gratefully acknowledge the support of H. U. Habermaier and his group, who provided their facilities and expertise for the sample preparation at the Max-Planck-Institut für Festkörperforschung, Stuttgart, Federal Republic of Germany, and of K. P. Frohmader, Institut für Elektrotechnik, Universität Erlangen, Federal Republic of Germany, for generating the masks.

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